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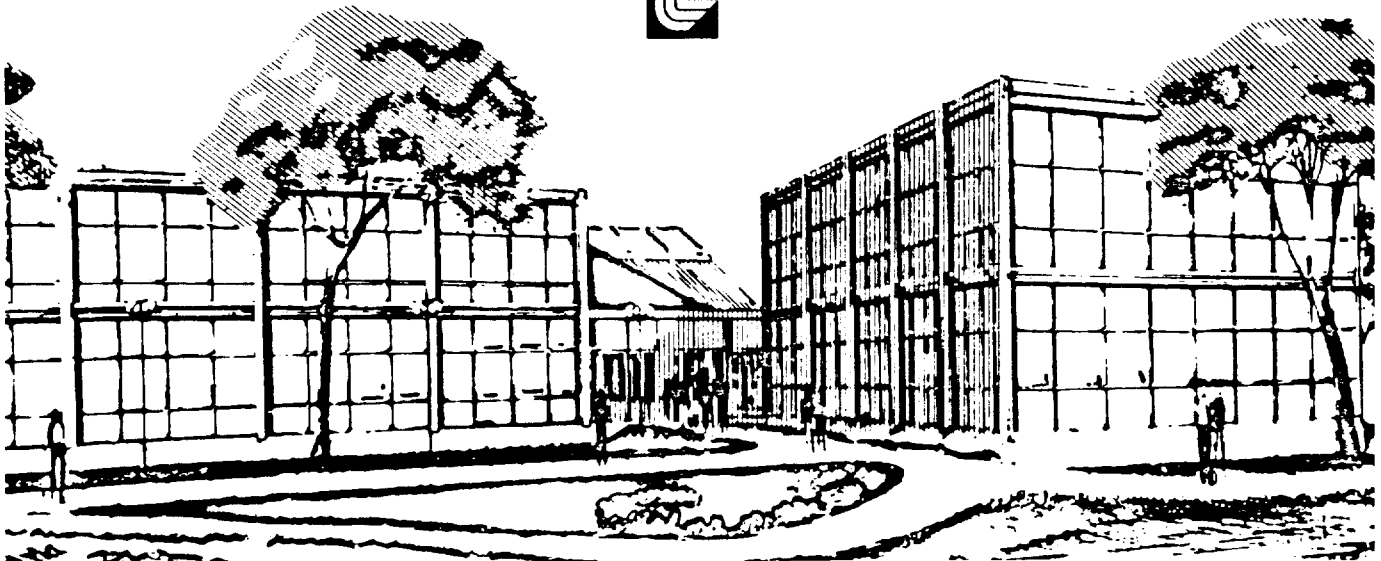
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A REVIEW OF THE MICROSCOPIC CROSS SECTIONS FOR THE AMERICIUM ISOTOPES IN THE RESOLVED RESONANCE REGION*

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ABSTRACT

The differential cross section measurements for ^{241}Am , $^{242\text{m}}\text{Am}$ and ^{243}Am are reviewed in the energy range from 0.5 eV to 10 keV. Parameters extracted from resonance analysis, such as the neutron strength function, the average level spacing, the average capture and fission widths, are compared for the various measurements. The average capture and fission cross sections from 100 eV to 10 keV are directly compared. The status of the data set is discussed with suggestions for further measurements.

I. INTRODUCTION

The Americium isotopes play an important role in the reactor production chain of the heavier actinides. They are pivotal in the formation of both ^{242}Cm and ^{244}Cm which present handling problems for fuel recycling and waste disposal schemes because of their high α -activity and their neutron emission from spontaneous fission and (α, n) reactions. The quantities of the heavier actinides that will result from uranium and plutonium recycling in nuclear power reactors have been estimated to be sizeable in the future [1]. A study was made by Hennelly [2] in which the adequacy of the actinide nuclear data was examined for the evaluation of heavy actinide production and removal programs. Although he concluded that the data was adequate at that time, he noted that large scale actinide burnup later in this century will require improved nuclear data because the expected large quantities of actinides could affect reactor design and operation.

This paper will review the status of the current differential cross section data for ^{241}Am , $^{242\text{m}}\text{Am}$ and ^{243}Am in the resolved resonance region with the aim of determining what additional measurements are required to produce an adequate set of nuclear data for these problems. The neutron energy range considered in this paper will be from 0.5 eV to 10 keV. A complete review of

the available integral and differential data for all the trans-actinides (including the Am isotopes) was made in 1975 and 1978 by Benjamin [3].

II. STATUS OF MICROSCOPIC DATA

A. ^{241}Am

There is now a considerable amount of differential data for ^{241}Am . For the total cross section, σ_T , Derrien and Lucas [4] have reported high-resolution measurements at the Saclay linac up to 150 eV neutron energy. Resonance parameters were extracted for 189 levels in this energy range. These data represent the best measurement of σ_T to date and all other data will be compared to them. Kalebin *et al.* [5] have measured σ_T for ^{241}Am from 0 to 30 eV while Belanova *et al.* [6] reported σ_T measurements from 8 to 30 eV. Both measurements were performed using a chopper at a reactor and the nominal resolutions of these measurements are a factor of 15 to 90 poorer than those of Derrien and Lucas. The only other σ_T data in the resonance range are the older measurements of Block *et al.* [7] and Slaughter *et al.* [8].

For the neutron absorption cross section, σ_a , the recent measurements of Weston and Todd [9] cover the neutron energy range from 0.01 eV to 370 keV. These data represent the most complete set for the absorption cross section in the resolved resonance range. Gayther and Thomas [10] have also recently measured σ_a for ^{241}Am in the energy range from 100 eV to 500 keV but these data were normalized to the data of Weston and Todd between 1 and 2 keV.

For the fission cross section, σ_f , there have been several new measurements. Derrien and Lucas [4] measured σ_f in the neutron energy range 0.8 to 150 eV by fission neutron detection. Knitter and Budtz-Jorgensen [11] have measured σ_f from 1 eV to several MeV using a new design fission fragment ionization chamber which discriminates against α -particle pileup effects. Gayther and Thomas [10] have also reported new measurements of σ_f from a few eV up to 20 keV using a fission neutron detector. Wisshak *et al.* [12] also have new data for σ_f from 10 keV to several MeV obtained by fission neutron detection. These new data are to be compared with the older σ_f data of Bowman *et al.* [13] which covered the energy range from 0.03 to 80 eV, the data of Gerasimov [14] covering 0.02 eV to 50 eV, the data of Seeger *et al.* [15] which covered 20 eV to 1 MeV, and the data of Shpak [16] which covered 8 keV to 3.5 MeV. Bowman and Gerasimov used spark counters at linacs to obtain their σ_f data while the Seeger data were obtained by fragment detection using an underground explosion as a neutron source. Shpak measured the fission fragments with the aid of passive glass track detectors.

The discussion of these data will be separated into two areas. The first will discuss the nuclear parameters obtained

from resonance analysis while the second will discuss the average cross sections from 100 eV to 10 keV. The results of the resonance analysis for all the measurements on ^{241}Am are summarized in Table I. The s-wave strength function, S_0 , extracted by Derrien and Lucas for the energy range 0-50 eV is $0.75 \pm 0.12 \times 10^{-4}$ which agrees well with the results of Kalebin et al. ($0.75 \pm 0.18 \times 10^{-4}$) in the 0-26 eV range and with the results of Belanova et al. ($0.75 \pm 0.23 \times 10^{-4}$) in the 8-30 eV range. For the expanded range 0-150 eV, however, Derrien and Lucas obtain a slighter higher value of S_0 equal to $0.94 \pm 0.09 \times 10^{-4}$. The strength function obtained by Slaughter et al. [8] from the Block et al. [7] OT data in the 0-45 eV range is $1.1 \pm 0.2 \times 10^{-4}$ which does not overlap the results of Derrien and Lucas. This discrepancy is apparently a result of three resonance groups at 6.74 eV, 9.17 eV and approximately 43 eV being assigned Γ_n values by Slaughter et al. which are an order of magnitude larger than those of Derrien and Lucas. For the average level spacing, \bar{D} , Derrien and Lucas obtain 0.55 ± 0.05 eV and Kalebin et al. obtain 0.60 ± 0.10 eV. Both results have been corrected for the effect of missed levels while the \bar{D} result of Belanova et al. (0.70 ± 0.10) has not been corrected.

The average s-wave capture width, $\bar{\Gamma}_\gamma$, was extracted by Derrien and Lucas in the 0-50 eV range using $\Gamma_\gamma^i = \Gamma_i - 2g\Gamma_n^0$ for many of the individual resonances. This can be done with good accuracy since both Γ_n and Γ_f are small compared to Γ . The average value was obtained from $\bar{\Gamma}_\gamma = \bar{\Gamma}_\gamma^i - \bar{\Gamma}_f$ where a contribution for the average fission width ($\bar{\Gamma}_f = 0.230$ meV) was subtracted to account for the fission widths. (The average fission width is discussed below). Weston and Todd [9] also extracted values of $\bar{\Gamma}_\gamma$ for six levels by fitting their σ_a data with single-level Breit-Wigner shapes. The average of these eight values is 47.1 ± 1.8 meV which is higher than the value of 43.77 ± 0.72 meV obtained by Derrien and Lucas. But since Γ_γ is large compared to Γ_n and Γ_f , the absorption cross section and the total cross section are almost equivalent. Therefore, the resonance area in absorption is not sensitive to Γ_γ and it is difficult to obtain an accurate $\bar{\Gamma}_\gamma$ value from shape analysis. The difference between the two values can be expected and the more accurate value of Derrien and Lucas should be preferred. The most complete set of fission widths was also extracted by Derrien and Lucas from their σ_f measurement in the 0-40 eV range. Γ_f values for thirty-eight (38) resonances were obtained from which an average value of 0.230 meV was derived. The distribution of fission widths was consistent with a χ^2 distribution of four (4) degrees of freedom which implies a large number of "open" fission channels. Derrien and Lucas also analyzed the resonance data of Seeger et al. [15] in the 20 to 50 eV range which resulted in a distribution of fission widths consistent with a χ^2 distribution of 15 degrees of freedom and an average Γ_f equal to 0.52 meV. This discrepancy was felt to be a result of either a normalization problem with the LASL data or a contamination of the fission events by capture events. The resonance parameters of Bowman et al. [13] in the 0-15 eV range

are in reasonable agreement with Derrien and Lucas. Derrien and Lucas noted that there were four resonances (at 3.97, 4.97, 6.12 and 9.11 eV) for which Bowman et al. reported Γ_f values 10 times smaller than their results. However, Table III of Ref. 13 was revised by Bowman et al. to correct for typographical errors in which the Γ_f values at 3.97, 4.97, and 6.12 eV were off by exactly a factor of 10. However, the resonance at 9.11 eV was not listed incorrectly but in this case the results of Bowman et al. are only a factor of 3 less than Derrien. With the revised set of Bowman parameters for 10 levels between 0 and 6.5 eV, one calculates an average $\bar{\Gamma}_f$ of 0.25 meV. From the first ten levels reported by Gerasimov [14], an average fission width of 0.22 meV is calculated. Knitter and Budtz-Jorgensen [11] analyzed 10 resonances in the 1-15 eV range missing four of the weaker resonances seen by Derrien and Lucas in this region. In general the results agree well with those of Derrien and Lucas. Gayther and Thomas [10] analyzed 12 fission resonances in the 1 to 15 eV range and the fission widths are also in good agreement with Derrien and Lucas.

In summary, the resonance parameters of Derrien and Lucas represent the best set of data for ^{241}Am in the resolved resonance region. Since the fission measurement of Derrien was performed by fission neutron detection it would be useful to corroborate their average fission width and width distribution via fission fragment detection as attempted by Knitter and Budtz-Jorgensen. This latter measurement suffered from a ^{239}Pu contaminant in the ^{241}Am sample and a new measurement using a cleaner sample should be forthcoming. It would be useful if the resonance parameters of the new measurements cover at least the 1-40 eV region covered by Derrien and Lucas. In relation to the capture width, the precision of the Derrien and Lucas value would be difficult to improve upon significantly. The level spacing result also should be adequate for present purposes. It would be useful to determine if the neutron strength function value of $0.94 \pm 0.09 \times 10^{-4}$ obtained by Derrien and Lucas for the 0-150 eV is the best representative value for S_0 or whether more weight should be placed on the 0-50 eV value of $0.75 \pm 0.12 \times 10^{-4}$ since the quality of the data are much higher in this region.

For the energy region between 100 eV and 10 keV, the average absorption cross section data and average fission cross section data will be reviewed. Figure 1 shows the recent σ_a measurements of Weston and Todd [9] and of Gayther and Thomas [10] in this energy range. The Gayther and Thomas results were normalized to the Weston and Todd data between 1 and 2 keV. The Weston and Todd data were normalized in the 0.02 to 0.03 eV region to the 2200 m/sec cross section. It can be seen from Figure 1 that the two measurements agree very well in shape in this energy range within the quoted uncertainties. The values shown in Figure 1 have a $\pm 10\%$ uncertainty for the Weston and Todd data and $\pm 12\%$ uncertainty for the Gayther and Thomas data. The only point not within 1 σ overlap is the 150 to 200 eV point. It should be noted that Fig. 1

shows only the average absorption cross section. Weston and Todd point out that there is considerable resonance structure above 50 eV which could be important because of resonance self-protection effects.

Figure 2 shows the average fission cross section for ^{241}Am obtained by the various authors between 100 eV and 10 keV. It is now clear that the underground explosion results of Seeger et al. [15] are not correct as has been conjectured for some time. For the more recent data, Gayther and Thomas [10] appear to be systematically higher than Knitter and Budtz-Jorgensen [11] particularly in the 1 to 10 keV region where up to a factor of 2 can be found. Both measurements were corrected for a contribution from ^{239}Pu impurities in the sample. The Gayther and Thomas data were measured using fission neutron detection with an $^{241}\text{AmO}_2$ sample and had to be corrected for a large neutron background from $^{17,18}\text{O}(\alpha, n)$ reactions. The Knitter and Budtz-Jorgensen data were obtained by fission fragment detection. Both measurements were made relative to the ^{235}U fission cross section so that the discrepancy cannot be accounted for by a normalization error of that magnitude. There is also a shape difference between the two measurements. The measurement of Shpak [16] using glass track detectors with a monoenergetic neutron source provides a point near 8 keV which is in good agreement with the Knitter-Budtz-Jorgensen data. Also the measurement of Wisshak et al. [12] provides a preliminary result at 11.3 ± 2.9 keV which overlaps the present data set and also agrees with the Knitter-Budtz-Jorgensen data. The accuracy of either of the two white source σ_f measurements (± 20 -25%) is probably sufficient for present purposes. However, the disagreement in the absolute value and the shape should be resolved by further measurements which do not suffer from the ^{239}Pu contaminants or the (α, n) neutron background. Both problems can be eliminated without significant difficulty.

In summary, the ^{241}Am differential cross section data now form a reasonably complete and consistent set in the energy range from 0.5 eV to 10 keV. Some improvements can be made in the fission data but the major anomaly has been removed. Another very important measurement for the future is a differential measurement of the branching ratio for neutron capture to the 152 yr isomeric state of ^{242}Am and to the 16 hr ground state. This branching ratio directly affects ^{242}Cm and ^{244}Cm production since the isomeric state of ^{242}Am is sufficiently long-lived to result either in ^{243}Am production through neutron capture or in burnup through fission. There are plans for a future differential measurement of this branching ratio at Karlsruhe [17].

B. ^{242}Am

The ground state of ^{242}Am is relatively short-lived (16 hr) compared to the first excited state (152 yr) so that its cross sections are less important for actinide production and burnup.

This section will cover only differential measurements on the isomeric first excited state, ^{242m}Am .

The only differential cross section data to date on this nucleus consist of three fission cross-section measurements. Bowman et al. [18] measured σ_f from 0.02 eV to 6 MeV using a spark chamber with a sample which consisted of 20% ^{242m}Am . Seeger et al. [15] measured σ_f from 20 eV to 1 MeV using an underground explosion as a neutron source. These data were taken concurrently with the ^{241}Am data mentioned in the previous section. The purity of the sample was basically the same as that used by Bowman et al. (i.e. 20% ^{242m}Am , 80% ^{241}Am). Recently Browne et al. [19] obtained new data covering the energy range from 0.01 eV to 20 MeV. These data were taken using a hemispherical fission chamber and a sample whose purity was 99.2% ^{242m}Am . Bowman et al. extracted resonance parameters for the first six resonances using shape analysis with a sum of single-level Breit-Wigner resonances. The s-wave strength function was reported to be $1.8 \pm 1.1 \times 10^{-4}$ per spin state. The level spacing was calculated to be $\bar{D} = 0.6 \pm 0.2$ eV but was not corrected for missed levels. The average fission width obtained was 460 meV and the number of "open" fission channels was estimated from the Bohr-Wheeler relationship $(2\pi\bar{\Gamma}_f/\bar{D})$ to be five (5). Browne et al. fit 45 levels below 20 eV using a sum of single-level Breit-Wigner resonances. An s-wave strength function equal to $1.25 \pm 0.15 \times 10^{-4}$ was extracted. The level spacing was calculated to be 0.38 ± 0.05 eV correcting for 15% missed levels. The average fission width from these data was 385 meV and the distribution of fission widths was analyzed with the maximum likelihood technique and found to be consistent with a χ^2 distribution of 10 degrees of freedom. In both the Bowman et al. and Browne et al. analysis, a sum of single-levels was found to be adequate because the large number of open fission channels reduced channel-channel interference and hence the need for a multi-level R-matrix fit. In the region of overlap the resonance parameters were found to be consistent between the two measurements. The data of Seeger et al. [15] were not analyzed for resonance parameters but since these data were obtained in the same measurement as the ^{241}Am data it is possible that the effect noted by Derrien regarding either normalization problems or contamination of the fission events by capture events is present in the ^{242m}Am data as well.

The average fission cross section results for ^{242m}Am are shown in Figure 3 between 100 eV and 10 keV. It can be seen that the data of Bowman et al. are consistently higher than the Browne et al. data particularly above 300 eV. The discrepancy varies from 20% to 50%. The Bowman data were measured relative to ^{239}Pu above 1 keV and the shape used for this reference cross section appears to differ significantly from more recent results. The normalization of the low energy Bowman data (referenced to the 2200 m/sec value) to this higher energy data is statistically limited to $\pm 10\%$. This uncertainty and the ^{239}Pu shape uncertainty

may account for the discrepancy above 1 keV but does not explain the discrepancy below 1 keV.

A high-resolution measurement of the total cross section would be very valuable to check the Γ_f values and the neutron strength function derived from the σ_T data. Since $\Gamma_f \approx \Gamma$, the total cross section and the fission cross section essentially will be equivalent. With current experimental techniques, it is not possible to measure the differential capture cross section for ^{242m}Am in the resolved resonance range. This is due to the 0.5% α -decay branch of ^{242m}Am to ^{238}Np which subsequently β^- decays to a group of excited states in ^{238}Pu resulting in ~ 1 MeV γ -ray emission to the ^{238}Pu ground state. Any capture cross-section measurement will have a background of 10^9 1 MeV γ -rays sec.gram suppress. The only current experimental technique that could deal with such a background is associated with using an underground explosion as a neutron source. Since these are extremely expensive and difficult, such a measurement may not be forthcoming in the near future. One indirect solution may be an improved thermal capture cross section value which presently has a large uncertainty (1400 ± 860 b). Since the thermal cross section is dominated by a large resonance at 0.173 eV, it may be possible to extract a Γ_γ for this resonance from the thermal capture cross section. This determination of Γ_γ would permit more accurate calculations of the capture cross section in the resolved resonance region.

C. ^{243}Am

Most of the differential cross section data for ^{243}Am in the resolved resonance region consist of total cross section measurements although there was one fission cross section measurement using an underground explosion. The best measurement of σ_T was reported by Simpson et al. [20] which covers the neutron energy range from 0.5 eV to 1 keV. Belanova et al. [21] performed σ_T measurements from 0.4 to 34 eV using a reactor chopper whose resolution was nominally two orders of magnitude poorer than obtained by Simpson et al. Berreth and Simpson [22] also performed σ_T measurements in the 0-25 eV range using a fast chopper. Cote' et al. [23] performed the earliest σ_T measurements in the 0-16 eV range using a fast chopper. The results of the resonance analysis performed in the above experiments are summarized in Table II. Since the results of Simpson et al. have the largest statistical sample and the best resolution, they provide the best information. The s-wave neutron strength function obtained by Simpson et al. for the energy range 0.5 to 250 eV is $S_0 = 0.96 \pm 0.10 \times 10^{-4}$. The results of Belanova et al. ($S_0 = 0.89 \pm 0.21 \times 10^{-4}$) and Cote' et al. ($S_0 = 0.84 \pm 0.25 \times 10^{-4}$) are consistent with this measurement. The average level spacing was extracted by Simpson et al. for the energy range 0-50 eV to be 0.68 ± 0.06 eV. This result was corrected for missed levels. Belanova et al. obtained $D = 0.71 \pm 0.06$ while the Berreth and Simpson results are consistent with $D = 0.68$. The capture width was extracted for 24 resonances below 18 eV by

Simpson et al. from which an average value of $\bar{\Gamma}_\gamma = 39 \pm 1$ meV was obtained. Berreth and Simpson list Γ_γ values for the first seven positive energy resonances from which an average value of 42 meV is obtained. Cote et al. obtained $\bar{\Gamma}_\gamma = 42 \pm 3$ meV from a weighted average of the first three resonances in their measurement. While the results of all the measurements are consistent, it is clear that the Simpson et al. data provide the best information to date.

The only differential fission cross section measurement to date was reported by Seeger et al. [24] for the energy range 50 eV to 3 MeV. No resonance analysis was performed on these data. The average fission cross section between 100 eV and 10 keV is shown in Figure 4. There appears to be an anomalous shape between 500 eV and 5 keV. There were experimental difficulties with this underground explosion measurement in the 10 keV to 100 keV range which could persist to lower energies. No other differential measurements exist to compare with at present.

It would be useful to have σ_f data over the 0.5 eV to 10 keV range to determine if there is a significant fission cross section to affect the production and burnup calculations and to check the Seeger σ_f shape in the keV range. A measurement of the absorption cross section for ^{243}Am similar to the Weston-Todd or Gayther-Thomas experiments would be valuable.

III. Summary and Conclusions

The differential cross section data for ^{241}Am now appear to be in reasonable agreement for the energy range 0.5 eV to 10 keV. A minor discrepancy remains for the absolute value and shape of σ_f between the Knitter-Budtz-Jorgensen data and Gayther-Thomas data, but this may be resolved in the near future with improved measurements. It is not clear whether the s-wave strength function of Derrien and Lucas in the 0-50 eV or 0-150 eV range is to be preferred. The respective values of S_0 are $0.75 \pm 0.12 \times 10^{-4}$ and $0.94 \pm 0.09 \times 10^{-4}$ which barely overlap. An evaluation of the quality of the data in the expanded energy range should be made to determine if this provides the more representative value for S_0 as might be expected for statistical reasons.

There are only two differential measurements of σ_f for ^{242m}Am that should be compared. The resonance parameters of Bowman et al. and Browne et al. agree within the very limited range of overlap (0-3 eV). However, there are very significant differences in shape and magnitude in the 100 eV to 10 keV range. Although the recent results of Browne et al. were obtained with a high purity sample compared with the 20% pure sample of Bowman et al., additional measurements of σ_f would be valuable to clarify the situation. Since the σ_f data of Seeger et al. for ^{242m}Am were obtained with a low-purity sample on the same underground explosion as the discrepant ^{241}Am data, they should be considered suspect at this time. A high-resolution σ_f measurement in the

resolved resonance region would be valuable to compare with the resonance parameters of Browne et al. below 20 eV. A 5-10% measurement of the thermal capture cross section would be useful in estimating σ_c for calculations of the ^{242}Am capture cross section.

The σ_T data for ^{243}Am are in reasonable agreement so that no new measurements are required at this time. However, since there is only one σ_f measurement (underground explosion) and no σ_a measurements, both would be valuable for calculations related to ^{244}Cm production. Techniques developed for similar measurements on ^{241}Am are directly applicable so that it should not be too difficult to obtain these data.

In summary, the nuclear data for the Am isotopes are close to being adequate for solving the immediate actinide burnup and production questions. But improvements in several areas can be made with present technology to provide a data set that should be valuable for even long range problems.

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Table I. Average resonance parameters for ^{241}Am derived from total, fission, and absorption cross-section data.

Experimenters [Ref.]	Type of Measurement	Energy Range ^a (eV)	S_0 ($\times 10^4$)	\bar{D} (eV)	$\bar{\Gamma}_\gamma$ (meV)	$\bar{\Gamma}_f$ (meV)
Derrien and Lucas [4]	σ_T	0-50	0.75(0.12)	0.55(0.05) ^b	43.77(0.72)	-
	σ_T	0-150	0.94(0.09)	-	-	-
	σ_f	1-40	-	-	-	0.230
Kalebin <u>et al.</u> [5]	σ_T	0-26	0.76(0.18)	0.60(0.10) ^b	-	-
Belanova <u>et al.</u> [6]	σ_T	8-30	0.75(0.23)	0.70(0.10)	-	-
Weston and Todd [9]	σ_a	8-50	-	-	47.1(1.8) ^c	-
Bowman <u>et al.</u> [13]	σ_f	0-15	-	-	-	0.25 ^d
Gerasimov [14]	σ_f	0-15	-	-	-	0.22
Gayther and Thomas [10]	σ_f	1-15	-	-	-	0.29 ^e
Knitter and Budtz-Jorgensen [11]	σ_f	1-15	-	-	-	0.34 ^e
<p>a. Energy range refers to region over which resonance parameters were extracted, not necessarily the energy range of the complete measurement.</p> <p>b. Corrected for missed levels.</p> <p>c. Calculated from first six positive energy resonances in Table I of Ref. 9.</p> <p>d. Calculated from revised resonance parameters for levels below 6.5 eV in Table III of Ref. 13.</p> <p>e. Calculated from resonance parameters in Table 5 of Ref. 11.</p>						

Table II. Average resonance parameters for ^{243}Am
derived from total cross-section data

Experimenters [Ref.]	Energy ^a Range (eV)	S_0 ($\times 10^4$)	\bar{D} (eV)	$\bar{\Gamma}_\gamma$
Simpson <u>et al.</u> [20]	0.5-250 0-50	0.96(0.10) -	- 0.68(0.06) ^b	- 39(1)
Belanova <u>et al.</u> [21]	0.4-35	0.89(0.21)	0.71(0.06)	-
Berreth and Simpson [22]	0-25	-	0.68	42
Cote' <u>et al.</u> [23]	0-16	0.87(0.25)	-	42(3)

^aEnergy range refers to region over which resonance parameters were extracted, not necessarily the energy range of the complete measurement.

^bCorrected for missed levels.

FIGURE CAPTIONS

1. The average absorption cross section for ^{241}Am from 100 eV to 10 keV.
2. The average fission cross section for ^{241}Am from 100 eV to 10 keV.
3. The average fission cross section for $^{242\text{m}}\text{Am}$ from 100 eV to 10 keV
4. The average fission cross section for ^{243}Am from 100 eV to 10 keV.

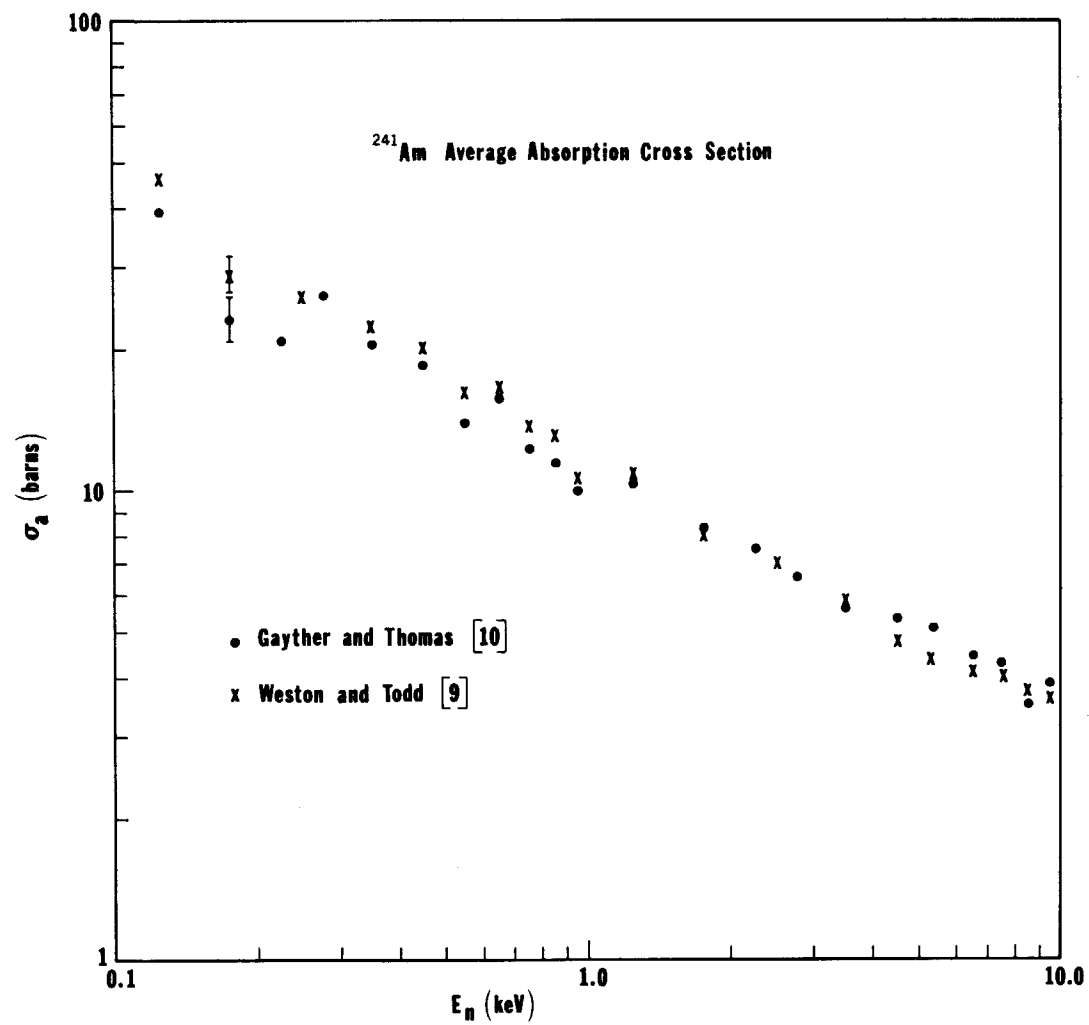


Fig. 1

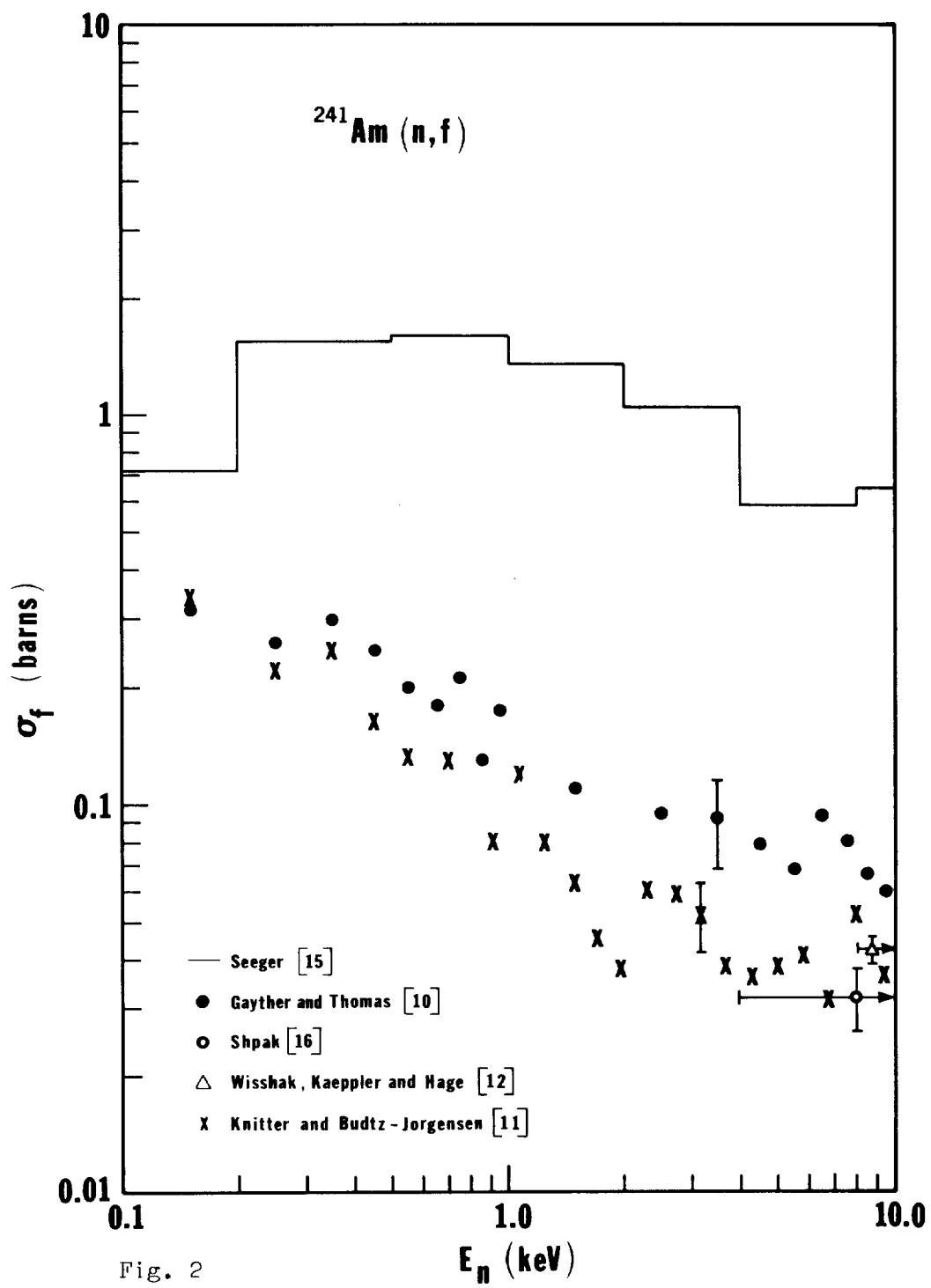


Fig. 2

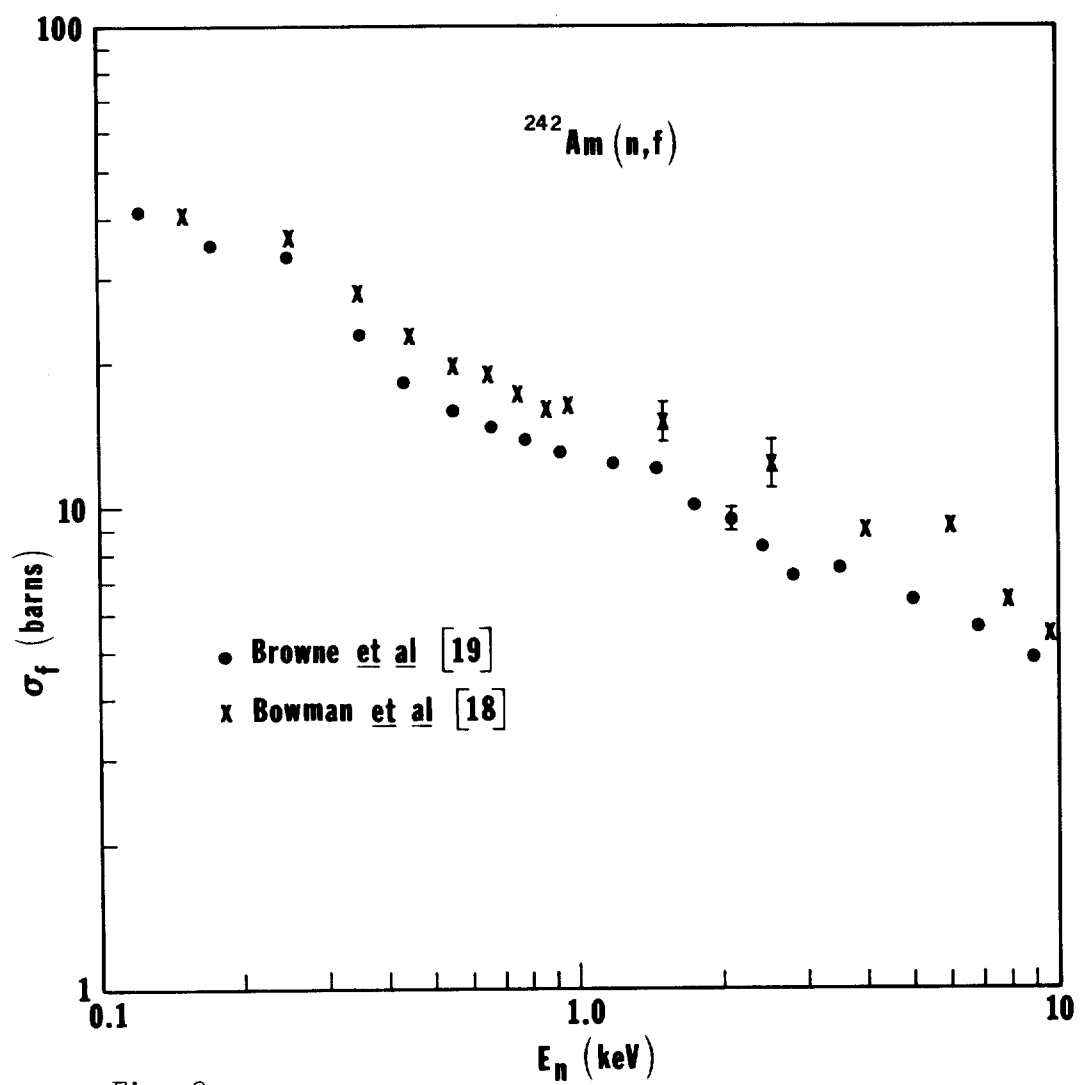


Fig. 3

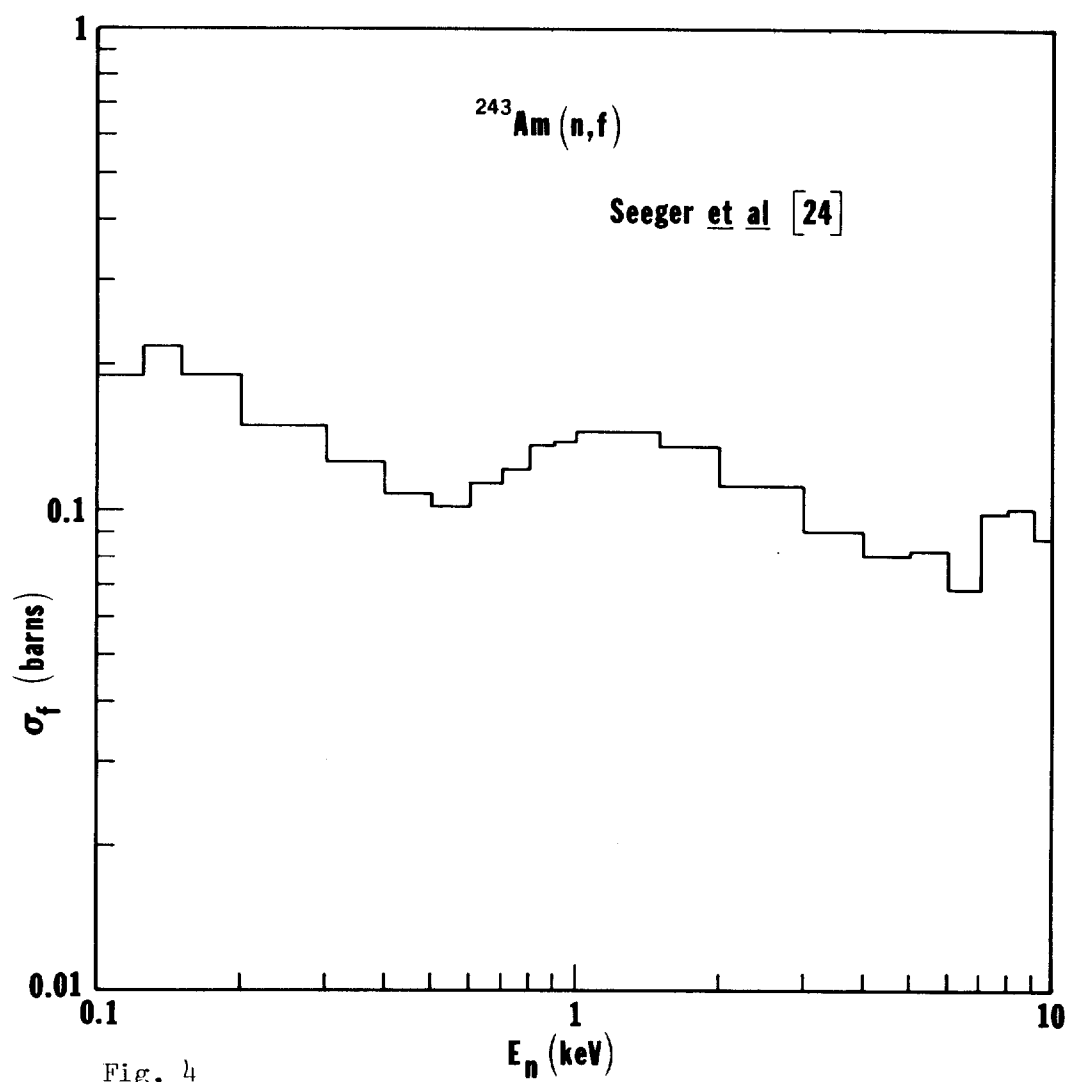


Fig. 4